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Mixing by shear, dilation, swap, and diffusion

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Abstract

This paper presents a theory of poroviscosity for binary solutions. Subject to mechanical forces and connected to reservoirs of molecules, a binary solution evolves by concurrent flow and diffusion. Our theory generalizes the classical theory of interdiffusion by decoupling the molecular processes for flow and diffusion. We further remove the assumption of local chemical equilibrium, so that the insertion of molecular into a material element, accompanied by a change in volume, is treated as nonequilibrium process and is put on the same footing as the process of shear deformation by viscous flow. The theory of poroviscosity has an intrinsic length scale, called the poroviscous length, below which the homogenization of a composition heterogeneity is limited by viscous flow, rather than by diffusion. The theory has implications for the analysis of interdiffusion in systems that display a decoupling between flow and diffusion, such as supercooled liquids, glasses, and physical gels. We illustrate the theory with numerical examples of a layered structure and a spherical particle. We discuss the results for feature sizes below and above the poroviscous length.

Keywords: Interdiffusion; Creep; Supercooled liquids; Glasses; Gels.

1. Introduction

Diffusion and flow in liquids and solids require molecules to change neighbors. Diffusion refers to the migration of a particle relative to its surrounding molecules, and the diffusivity D characterizes the rate of diffusion. Flow refers to the molecular motion to relax an applied stress, and the viscosity η gives the proportionality ratio between the stress and strain rate. Einstein (1905) assumed that a particle diffuses in a liquid by the Stokes creep of the liquid, and related the diffusivity D of the particle to the viscosity η of the liquid:

$$\frac{D\eta}{kT} = \frac{1}{Ca} \quad (1)$$

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